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Electrical properties of bipolar plate and gas diffusion layer in PEFC

Atsushi Miyazawa [a,b,*], Tomokatsu Himeno [b], Atsushi Nishikata [a]

[a] Department of Chemistry and Materials Science, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8552, Japan
[b] Nissan Research Center, Nissan Motor Co. Ltd., 1 Natsushima-cho, Yokosuka, Kanagawa 237-8523, Japan

HIGHLIGHTS

► No noticeable increase in contact area with increasing compression pressure above a level of 0.8 MPa.
► At a compression pressure of 2.0 MPa, contact area ratio of 38% was obtained.
► The decrease in contact resistance relative to Au coverage was less than 1 mΩ cm^2 over coverage of 0.6.
► Contact resistance depends not only on the coating coverage but on in-plane resistance of the GDL.

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ABSTRACT

This study investigated the electrical properties of the gas diffusion layer (GDL) and the bipolar plates that are key components of a polymer electrolyte fuel cell (PEFC). Observations of the contact condition between the GDL and a bipolar plate under compression showed no noticeable increase in contact area with increasing compression pressure above a level of 0.8 MPa. Contact resistance between the GDL and an Au-coated bipolar plate decreased with increasing Au coverage. However, the decrease in contact resistance relative to Au coverage was small, less than 1 mΩ cm^2, at coverage of approximately 0.6 or higher. That is presumably attributable to the flow of electrons to the Au contact spot with little voltage loss in the in-plane direction of the GDL. One reason for that is the fact that sufficient contact with the GDL was secured, as indicated by the observation results. Additionally, the in-plane resistance of the GDL was markedly smaller than the contact resistance. The change in contact resistance relative to Au coverage was calculated for GDLs having different in-plane resistivities. The results revealed that the increase in contact resistance relative to Au coverage became smaller as the resistivity of the GDL decreased.

1. Introduction

Reducing the cost of the fuel cell stack is one of the most critical issues that must be addressed to commercialize fuel cell vehicles (FCVs). One cost reduction approach that can be considered is to substitute lower-cost materials used to fabricate FC stack components. Another effective way is to downsize the fuel cell stack itself by improving its power density. Reducing the electrical resistance is a markedly effective way for automotive fuel cells that operate in high current density condition. Lowering the contact resistance at the interfaces between stacked components in particular contributes substantially to higher output. For the purpose of reducing the contact resistance between the gas diffusion layer (GDL) and the bipolar plates, researchers have examined the base metal of the plates and surface treatment materials [1–5]. Regarding the GDL, on the other hand, because carbon fiber is an excellent electric conductor, studies have been reported concerning gas diffusion, removal of product water and heat transfer properties [6–8], but there are very few reports about the electrical resistance of the GDL itself [9–11]. In addition, the details of the contact condition at the interface with the bipolar plates have not been clarified yet.

In actuality, investigations of bipolar plate materials alone, as done in studies conducted to date, are not sufficient for reducing both contact resistance and cost. It is necessary to address both of these issues on the basis of detailed information concerning the electrical resistance of the GDL itself and the contact condition between the GDL and the bipolar plates. Therefore, in this research, detailed investigations were made of the electrical resistance of both the GDL and the bipolar plates and their contact resistance. With regard to contact resistance in particular, observations were

* Corresponding author. Department of Chemistry and Materials Science, Tokyo Institute of Technology, 2-12-1, O-okayama, Meguro-ku, Tokyo 152-8552, Japan. Tel.: +81 46 867 5181; fax: +81 46 865 5796.
E-mail address: atsu-miyazawa@mail.nissan.co.jp (A. Miyazawa).

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Table 1
Specification of bipolar plate.

Sample	Base metal		Surface coating			
			Undercoat		Topcoat	
	Metal	Thickness (mm)	Metal	Thickness (nm)	Metal	Thickness (nm)
Ni–Au	SS316L	0.1	Ni	100	Au	0–100
D–Au	SS16L	0.1	–	–	Au	0–100

made of the contact condition of these two components, and the correlation between contact resistance and the compression condition, which is a key factor, was confirmed. Based on the results, measures were then considered for reducing both electrical resistance and cost.

2. Experimental

2.1. GDL and bipolar plates

Three types of GDL were prepared for examination using commercially available carbon paper TGP-H-30, -60 and -90 (Toray Industries, Inc., [12], denoted as H30, H60, H90 respectively). Their thickness is 110, 190 and 280 μm respectively. As shown in Table 1, the bipolar plates were made of SS316L austenitic stainless steel of 0.1 mm in thickness as the base metal. The surface treatment applied to SS316L was an Au electroplating, which is one of the best surface treatment materials on metallic bipolar plate. Samples were prepared of varying coating thickness from 0 to 100 nm. Two types of Au coating processes were used. In one process, an intermediate Ni layer was first plated on SS316L to a thickness of 100 nm (denoted as the Ni–Au sample in the table); in the other process, the Au coating was applied directly to SS316L without any intermediate layer (denoted as the D–Au sample in the table). Electroplating thicknesses of over 5 nm was measured with an X-ray fluorescence (XRF) coating thickness gauge. Below 5 nm, the coating thickness was adjusted by varying the current and coating time. Surface coverage was calculated on the basis of an AES surface analysis and image processing results.

2.2. Measurement of electrical resistance

The experimental setup used to measure the contact resistance between the GDL and a bipolar plate and the electrical resistance along the depth of the GDL under various compression pressures is shown in Fig. 1-(a). Resistance was measured with a 4-probe resistance system. A bipolar plate was sandwiched between two GDLs and a direct current (DC) 1 A was applied between two terminals. The voltage drop between the terminals was measured at the copper electrode having a diameter of 20 mm. Voltage was measured at the 1-mm-diameter terminal that was insulated from the terminal in the center of the copper electrodes. Based on the resistance obtained with the relation $R = V/I$, contact resistance (R_{cr}) was calculated as

$$R_{cr} = S_1(R_{all} - 2R_{GDL})/2 \tag{1}$$

where R_{all} denotes all the measured resistance, R_{GDL} is the through-plane resistance of the GDL, which was measured for one GDL sandwiched between the terminals, and S_1 indicates the area of the copper electrode.

Fig. 1-(b) shows the experimental setup used to measure the in-plane resistance of the GDL. Copper electrodes were plated to a thickness of 5 μm on a 5-mm-thick plastic plate, above which was positioned a 10-mm-wide GDL and compression pressure was applied. In-plane resistance is anisotropic in the transverse direction (TD) and in the machined direction (MD) of the GDL. The GDL was prepared two types of test piece to measure anisotropic resistance as shown in Fig. 1-(b).

A DC 100 mA was passed between the copper electrodes from both ends, and the resistance R' was measured for various voltage terminal lengths L. Resistivity ρ was calculated with the following equation,

$$\rho = R'S_2/L \tag{2}$$

Since the cross-sectional area S_2 of the GDL varied with the change in thickness due to the applied compression pressure, the load-displacement curve of the GDL was measured in advance. The average thickness found for the second curve cycle was used as the GDL thickness because hysteresis occurred in the initial loading–unloading cycle. Electrical resistance was measured to a maximum compression pressure of 4.0 MPa, taking into account the operating condition of automotive fuel cells.

2.3. Measurement of contact area on the GDL

Reports about the electrical resistance of GDLs concern the influence of their electrical properties on the power generation performance of a fuel cell and measurement of the electrical conductivity of the carbon fibers composing GDLs. Zhou and Liu used a model to calculate the power generation performance when several GDLs having different through-plane and in-plane electrical resistances were used. They reported that through-plane resistance directly influences the resistance overvoltage of a fuel cell and that in-plane resistance influences not only the overvoltage but also the in-plane distribution of the local current density in a cell [10].

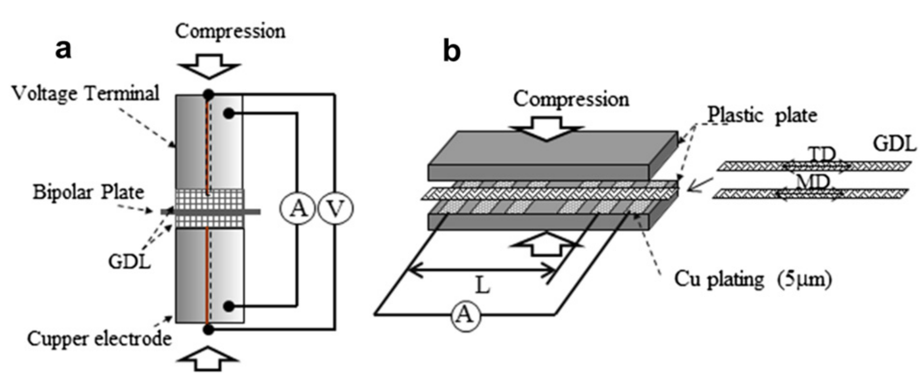

Fig. 1. The schematic configuration of electric resistance measurement. (a) Contact resistance and through-plane resistance, (b) in-plane resistance.

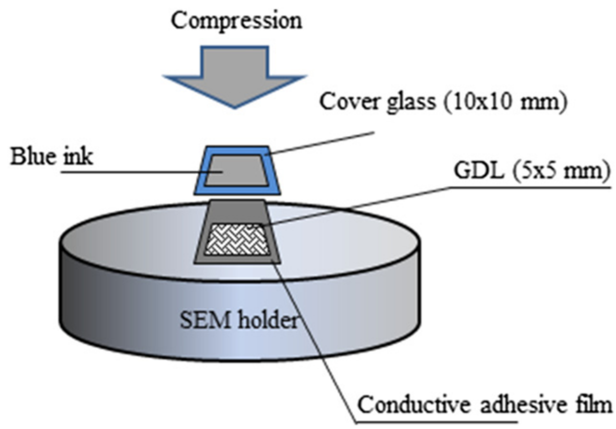

Fig. 2. Schematic diagram of GDL compression and SEM analysis.

Parikh et al. used an in situ 4-probe method inside a SEM to measure the electrical resistance of carbon fibers [11]. They reported a measured value of around 60 Ω and that the Kelvin resistance was reduced to approximately one-half through bending deformation process. However, not only are actual GDLs composed of multiple fibers, they also have a porous structure formed by the use of many graphitized binders in the in-plane direction to secure the fibers. Therefore, it is presumed that the anisotropy of the electrical properties of the GDL and the contact condition with the bipolar plates affect the contact resistance.

In this study, two investigations were carried out to confirm the contact condition between the GDL and the bipolar plates. One investigation was conducted to examine the carbon fiber part of the actual top layer of the GDL that is in contact with a bipolar plate. The other investigation involved observation of the contact condition between the GDL and a bipolar plate under the application of compression pressure. The carbon fiber part of the top surface layer of the GDL was observed with the aid of an optical microscope after impregnating resin inside the GDL by vacuum process. The images thus obtained were binarized with respect to the carbon fibers and the resin, and the area ratio of the carbon fiber part was calculated.

The experimental setup used to observe the contact condition between the GDL and a bipolar plate under compression pressure is shown schematically in Fig. 2. The GDL was fixed to the SEM holder and a glass plate simulating a bipolar plate was positioned above it. The contact condition between the GDL and the glass plate was observed while applying compression pressure to the latter. Before the observation was made, the glass plate was coated in advance with a blue ink consisting mainly of phosphotungstic acid, hexane and ethanol in order to adjust the wettability with the carbon fiber surface. The thickness of the ink layer was set at 0.3 μm, which was sufficiently thinner than the carbon fiber diameter (8 μm). The contact area was binarized using an image processing technique and the area of contact covered with the blue ink was calculated.

3. Results and discussion

3.1. Contact resistance between GDL and bipolar plate

Fig. 3 shows the through-plane resistance of the GDL and the contact resistance with the bipolar plate, which were measured under various levels of compression pressure from 0.2 to 4.0 MPa. The bipolar plate surface was coated with Au to a thickness of 108 nm that ensured sufficient coverage. The results indicate that the through-plane resistance differed substantially among the

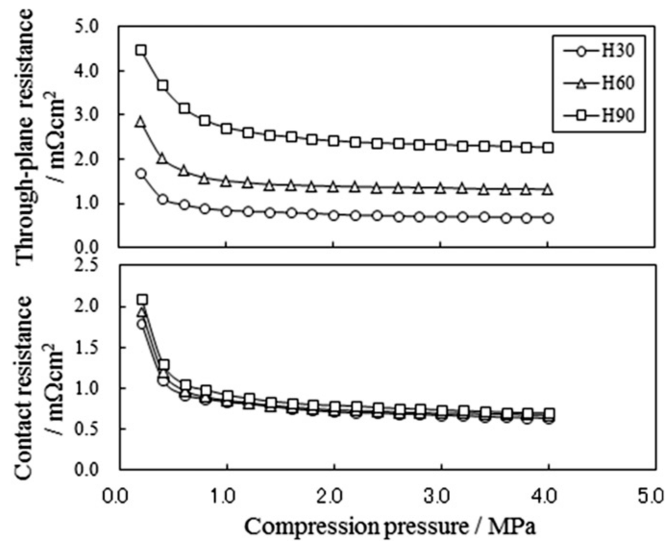

Fig. 3. Plots of through-plane and contact resistance of GDL against compression pressure.

three GDL specifications regardless of the compression pressure. The difference in resistance values corresponds to the difference in the thickness of the GDL. It is known that the resistance of the GDL, R_{GDL}, follows resistivity ρ_y in the depth (L) direction according to the following equation.

$$R_{GDL} = \int_{GDL} \rho_y \mathrm{d}y \approx \rho_y L \tag{3}$$

Contact resistance, on the other hand, was nearly the same regardless of the GDL specifications, and the values were smaller than the through-plane resistance. As indicated by the relation in Eq. (3) above, when the thickness of the GDL was reduced to that of H30, the results in Fig. 3 show that the through-plane resistance decreased to a level equal to the contact resistance. These results imply that, as a specific measure for reducing the resistance overvoltage of the GDL, making the GDL thinner so as to lower the through-plane resistance is a more effective approach than reducing the contact resistance.

3.2. Contact area on the GDL

Fig. 4 shows a microscopic image of the observed carbon fibers of the top surface layer of the GDL (H60) when impregnated with resin and a corresponding binary image obtained with an image processing technique. The area ratio of the fibers calculated by means of image processing was 23%. This value was close to the coverage data released by Toray for this GDL (H60) specification (i.e., porosity of 78% is equivalent to carbon area ratio of 22% on surface of the GDL.).

A SEM image obtained under compression pressure is shown in Fig. 5 along with a corresponding binary image obtained by image processing. At a compression pressure of 0.3 MPa, the top surface layer and some of the fibers underneath were in contact with the plate and the contact ratio was 9%. At a compression pressure of 0.6 MPa, the contact area markedly increased, as the fibers from the top surface layer to the second and third layers as well as the carbon parts connecting the fibers were in contact with the plate. The contact ratio increased to 22%. At compression pressures of 0.8 MPa or higher, no pronounced increase was seen for fibers newly in contact with the bipolar plate in relation to the increase in the

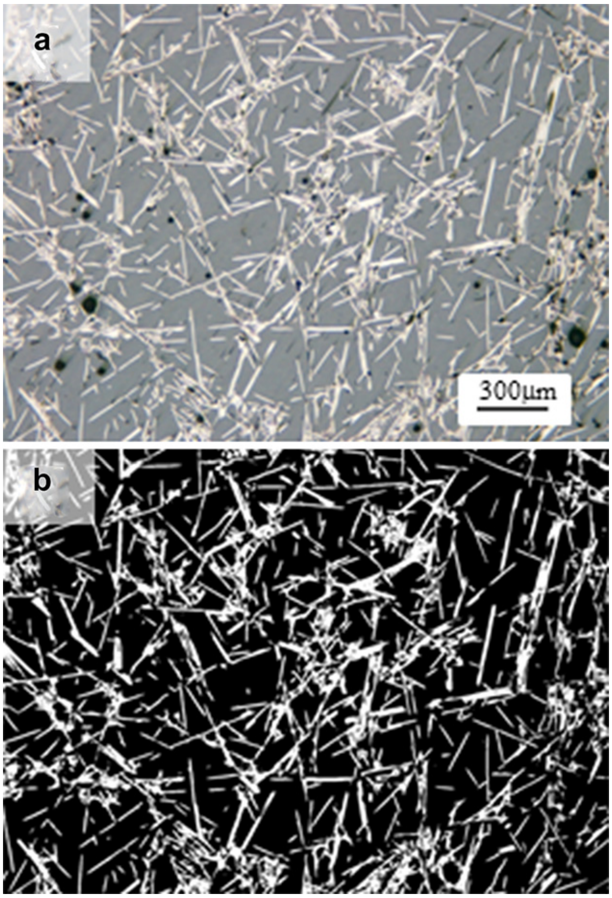

Fig. 4. Microscopic and binary images of the GDL (H60).

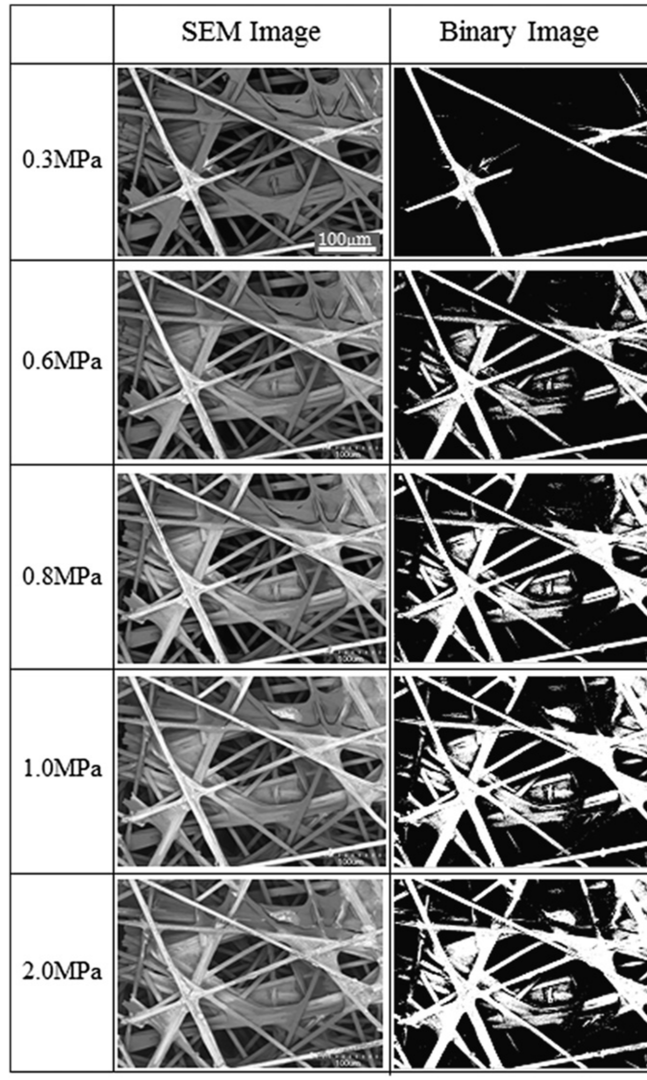

Fig. 5. SEM and binary images of GDL contact area under various compression pressures.

compression pressure. At a compression pressure of 2.0 MPa, a contact area ratio of 38% was obtained.

The contact area ratio was calculated from binary images like that in Fig. 5 and the results are plotted in Fig. 6 in relation to the compression pressure. As noted above, the area ratio of the carbon fiber part of the top surface layer was 23%. At a compression pressure of 0.6 MPa, a contact area of equal ratio was formed with the bipolar plate. At a compression pressure of 2.0 MPa, it is seen that a contact area ratio of 38% was obtained, which was 10% larger than the carbon fiber area of the top surface layer.

These results revealed that the change in the contact resistance between the GDL and the bipolar plate with increasing compression pressure was nearly identical to the change in the actual contact condition between the two. However, it was observed that the actual area of contact was larger than the area of the carbon fiber part of the top surface layer.

3.3. In-plane resistivity of the GDL

The in-plane electrical resistance of the GDL measured with the experimental setup in Fig. 1-(b) is shown in Fig. 7. Also shown is the through-plane resistance of the GDL that was converted to an equivalent resistivity. In-plane electrical properties are anisotropic in the TD and in the MD, with the resistivity in the latter direction being smaller than that in the former direction. At a compression pressure of 0.3 MPa, the resistivity in the transverse and machined directions was 7.8 and 4.5 mΩ cm, respectively. At 4.0 MPa, the corresponding values decreased slightly to 4.5 and 3.2 mΩ cm. This decrease in resistivity due to compression can presumably be attributed to increased contact between the carbon fibers composing the GDL and to reduce contact resistance between the carbon fibers. It is also seen that the in-plane resistivity was markedly smaller than the through-plane. The resistivity in the machined direction in particular was only 1/34 of the through-plane direction under compression pressure of 4.0 MPa.

3.4. Effect of bipolar plate surface treatment on contact resistance

In this study, the effect of the bipolar plate surface treatment on contact resistance with the GDL was also investigated. Two types of Au-coated bipolar plate samples were prepared. In one process, an intermediate Ni layer was coated on SS316L before applying the Au coating (Ni–Au sample); in the other process, the Au coating was applied directly to SS316L (D–Au sample). Fig. 8 shows the Au coverage as a function of the Au coating thickness. For both types of plate samples, coverage increased sharply with increasing Au coating thickness up to an Au thickness of 10 nm. At an Au coating thickness of 10 nm, both types of samples showed coverage of more than 90%. They both exhibited coverage of more than 98% at an Au coating thickness in the vicinity of 100 nm.

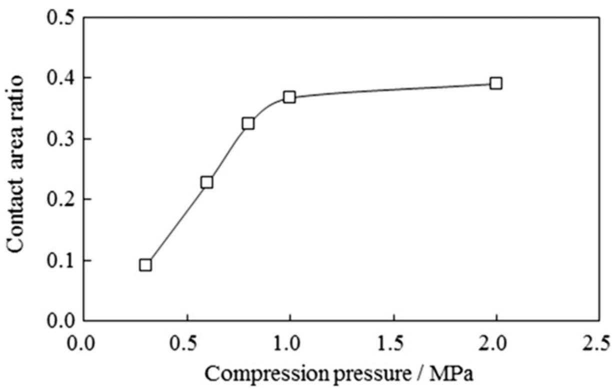

Fig. 6. Plots of contact area ratio under various compression pressures.

The change in contact resistance with the GDL in relation to the compression pressure is shown in Fig. 9-(a) and (b) for various coating thicknesses on Ni–Au and D–Au samples, respectively. In both figures, the 0-nm plots indicate the contact resistance with the Ni layer without any Au coating or with SS316L.

For the condition without any Au coating, the contact resistance between the GDL and SS316L was markedly high, showing a value of 44 mΩ cm^2 at a compression pressure of 4.0 MPa. Presumably, this high contact resistance can be attributed to the presence of an oxide film that formed on the surface of stainless steel. With an Au coating thickness of 1 nm (coverage of 68–78%), contact resistance declined noticeably; with a coating thickness of 10 nm or more (coverage $>$ 90%), contact resistance showed virtually no decline in relation to a further increase in the Au coating thickness.

The contact resistance for both types of coated samples at a compression pressure of 4.0 MPa is plotted as a function of Au coverage in Fig. 10. In the region of low Au coverage, the results show that contact resistance was higher for the D–Au type than for the Ni–Au type. That is probably due to the fact that a contact resistance component between the SS316L base metal and the GDL was included in the values of the D–Au type in addition to the contact resistance between Au and the GDL. Furthermore, seen from another point of view, the difference in contact resistance ascribable to the base metal in the low coverage region can also be interpreted as simulating a change in apparent resistance under a condition where the base metal corrodes in the fuel cell operating environment. This means that the magnitude of the contact resistance between the non-surface-treated portion of the bipolar plate

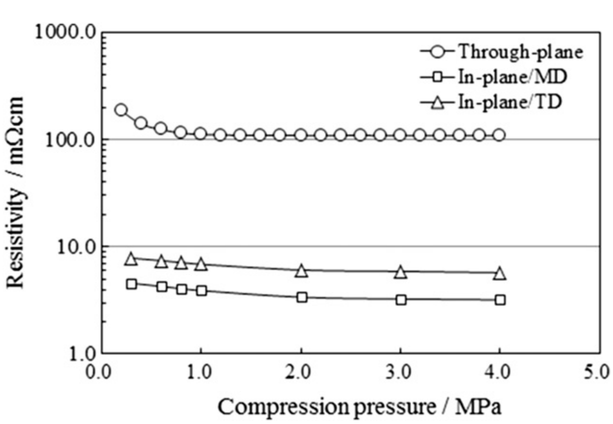

Fig. 7. Through-plane and in-plane resistance of the GDL under various compression pressures.

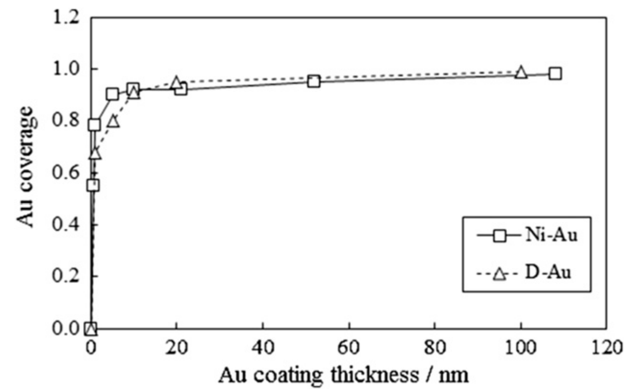

Fig. 8. Changes of Au coverage against Au plating thickness. (Ni–Au: SS316L/Ni/Au, D–Au: SS316L/Au).

surface and the GDL determines the lower allowable limit of the coverage obtained with the surface treatment process.

The results show, on the other hand, that there was virtually no difference in contact resistance between the two types of coating at Au coverage of 0.6 or higher. In addition, the increase in contact resistance relative to the increase in coverage was a markedly small value of less than 1 mΩ cm^2. The reason for that can be inferred as follows. As mentioned earlier, the in-plane resistance of the GDL is very small, so the in-plane flow of electrons in the GDL is controlled, thereby inhibiting an increase in contact resistance even if the plate surface treatment process produces coverage of 0.6.

This poses the question of how GDLs with different resistivities might influence the coverage obtained with the bipolar plate

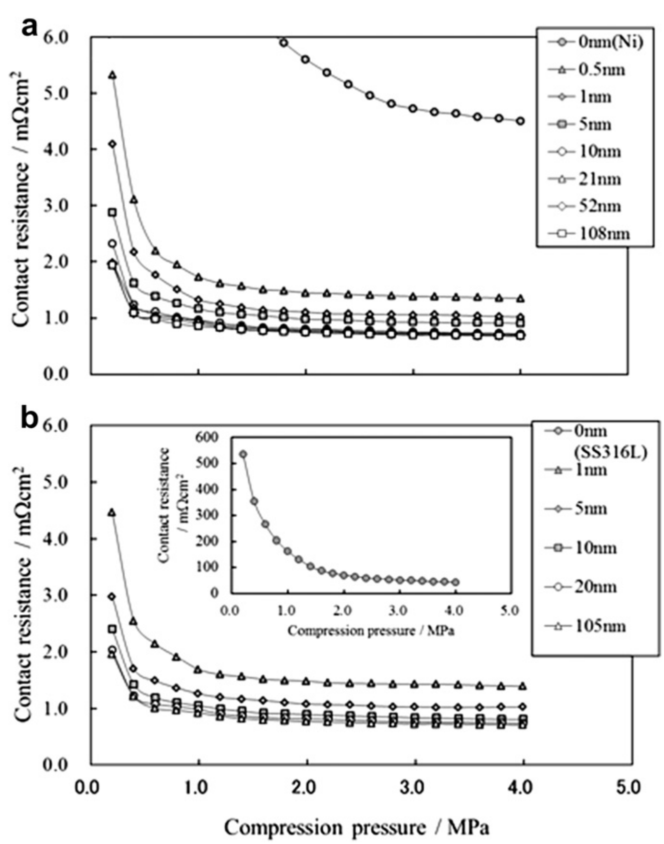

Fig. 9. Contact resistance between GDL and Au plated SS316L under various compression pressures. Au plating: (a) Ni–Au, (b) D–Au.

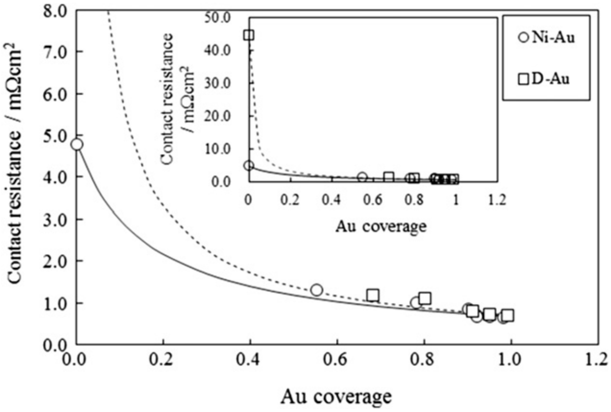

Fig. 10. Relationship between contact resistance and Au coverage.

surface treatment. That influence was investigated qualitatively using a relational equation for electrical contact. According to Holm, contact resistance (R_{cr}) can be expressed as the sum of constriction resistance R_c and film resistance R_f as shown in the well-known equation below [13].

$$R_{cr} = R_c + R_f = \rho/2a + \rho_f d/\pi a^2 \tag{4}$$

where a is the radius of contact spot, ρ is the resistivity of the material in contact and ρ_f is the film resistance. In this study, R_f (ρ_f) was ignored because contact was formed between Au and the GDL. Letting n represent the number of contact spots and ρ_1 and ρ_2 the resistivity of Au and the GDL, respectively, contact resistance can be expressed with the following equation.

$$R_c = (\rho_1 + \rho_2)\sum_n 1/2a_n \tag{5}$$

In general, electrical contact is considered to be formed between two opposing solid materials. In the present study, the GDL was porous and the contact area also changed due to the applied compression force. Nonetheless, it was assumed from the results in Fig. 6 that the carbon fibers were in sufficient contact under a compression pressure of 4.0 MPa. Letting $\rho_1 = 2$ μΩ cm, the change in R_c relative to the coverage was calculated for varying values of ρ_2, assuming a baseline value of $\rho_2 = 3.2$ mΩ cm from the results in Fig. 7.

Fig. 11 shows the calculated change in R_c in relation to ρ_2 normalized to a baseline of 1.0 for the coverage obtained with the surface treatment, when a GDL with a $\rho_2 = 3.2$ mΩ cm was used. The change in resistance as a function of Au coverage shows the same tendency as the measured in Fig. 10. The results indicate that R_c itself is also proportional to ρ_2. Moreover, it is also observed that the change in resistance relative to Au coverage differed among the GDL samples used. Specifically, the increase in R_c relative to the decline in Au coverage was smaller for GDLs with a lower ρ_2 value. The results imply that the coverage obtained with the bipolar plate surface treatment process can be reduced by controlling the in-plane conductivity of the GDL.

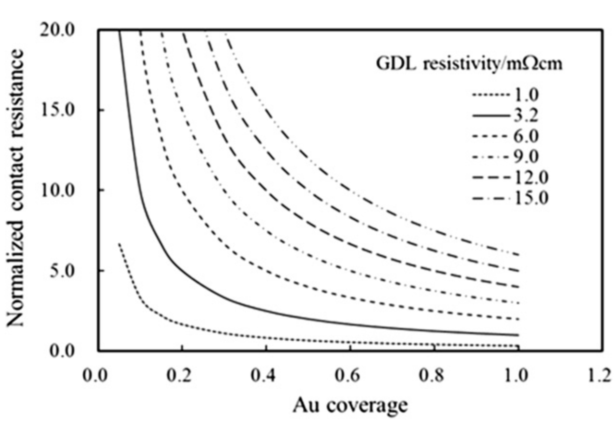

Fig. 11. Relationship between in-plane resistivity of GDL and Au coverage.

4. Conclusions

It was found that the contact resistance between an Au-coated bipolar plate and the GDL showed no difference among the H30, H60 and H90. However, the through-plane resistance of the GDL was proportional to the GDL thickness. For a GDL thickness of H30 or greater, the through-plane resistance was larger than the contact resistance between the bipolar plate and the GDL.

The contact resistance between the bipolar plate and the GDL decreased with increasing compression pressure. That tendency is attributed to an increase in the number of contact spots between the bipolar plate and the GDL with increasing compression force. However, at compression pressures of 0.8 MPa or higher, the contact spots with the bipolar plate are not limited to the fibers of the top surface layer of the GDL, but also include contact with the second and third internal layers of the GDL in some cases. That results in the formation of a larger contact area than that of the carbon fibers of the top surface layer.

The lower limit of the coverage of the Au coating formed on the bipolar plate surface is determined by the following two factors related to the contact resistance with the GDL at a compression pressure of 4.0 MPa. First, in the case of low Au coverage, contact resistance is influenced by the contact resistance between the GDL and the non-Au-coated portion of the bipolar plate. Consequently, the lower allowable limit of Au coverage is dependent on the contact resistance with the non-Au-coated portion. Second, at Au coverage of 0.6 or higher, sufficient contact is assured with the GDL and the in-plane conductivity of the GDL is also high. Accordingly, it is assumed that electrons flowing from the membrane electrolyte assembly tend to flow in the GDL in-plane direction to the Au contact area without any voltage loss. In other words, this suggests that the allowable coverage reduction obtained with the bipolar plate surface treatment for contributing to an overall cost reduction is dependent on the in-plane conductivity of the adjacent GDL.

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